DEFENCE R&D DÉFENSE

Radiological Air Sampling for the Canadian Forces

Final report for tasking 284761KROL (DSSPM)

D.S. Haslip, D. Estan
Defence Research Establishment Ottawa

Defence R&D Canada

DEFENCE RESEARCH ESTABLISHMENT OTTAWA

TECHNICAL MEMORANDUM
DREO TM 2001-033
June 2001

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Canada

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Radiation Effects Group
Space Systems and Technology Section

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Project
DSP 00002199

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Abstract

DREO has performed a study of air sampling methodologies and subsequent methods of data analysis. This has been done in support of Defence Services Procurement Project 00002199 (Radiation Detection, Identification, and Dosimetry) and is focussed on what methods would be best suited to the Canadian Forces. As part of this work, DREO performed a number of experimental trials to demonstrate and test the principles and methods of air sampling. A suggested method is presented for CF air sampling procedures. It uses high-volume air samplers, alpha-beta probes for detection of contaminants on the filter, and gamma-ray spectroscopy for additional sensitivity in some cases. Data analysis approaches are suggested, with appropriate caveats. One of the major findings of this study is that radon and thoron backgrounds present a serious impediment to the detection of low levels of airborne contaminants. In fact, the equipment discussed in this work is insufficient for the detection of alpha-emitting contaminants down to safe levels, even when the field-optimized methods of this study are employed. Reliance on these techniques in such scenarios risks radiation exposures.

Résumé

CRDO a étudié des méthodologies d'échantillonage d'air ainsi que des méthodes d'analyse de données suite à l'échantillonage. Nous avons effectué cette étude pour le projet 00002199 (détection, identification, et dosimétrie de rayonnement) tout en examinant quelles méthodes seraient meilleur pour les forces canadiennes. CRDO a effectué plusieurs essais expérimentales afin de démontrer les différentes méthodes d'échantillonage d'air. Nous avons suggéré une méthode que les forces canadiennes pourraient exécuter facilement. Cette méthode utilise les échantillonneurs d'air à haut volume, les sondes alpha-bêtas pour la détection des contaminants sur le filtre, et, dans certains cas, la spectroscopie de rayons gamma. Des méthodes d'analyse de données sont suggérées, ainsi que des suggestions en ce qui concerne la mise en oeuvre de ces méthodes. Un des résultats les plus importants de cette étude est sans aucun doute l'effet des produits radioactifs de radon et de thoron empêchant la détection de petites quantités de contaminants suspendus dans l'air. En fait, l'équipement dont nous discuteront dans ce document n'est pas adéquat pour la détection des contaminants alpha, même lorsque les méthodes décrites dans cette étude sont utilisées. Dans de telles situations, si trop de confiance est placée sur les techniques décrites dans ce document, il existe un risqué réel d'exposition à de niveaux élevés de rayonnement.

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Executive summary

Introduction: Defence Services Procurement Project 00002199 (Radiation Detection, Identification and Dosimetry) will be procuring air sampling equipment for the Canadian Forces. The project has contracted with DREO to study methodologies for air sampling and data analysis. This is the final report of this study, which included an extensive literature search and a series of experimental trials to demonstrate and test the principles elaborated herein.

Results: This paper outlines a protocol for air sampling and data analysis by the Canadian Forces, which could be easily implemented in the field. It has also described the shortcomings of this method, which centre on the radioactive background signal from radon and thoron decay products. Methods to eliminate this background are prone to error and are extremely sensitive to statistical fluctuations in the data. As a result, this background defines the sensitivity of this method to low-level airborne contaminants.

Significance and Future Plans: The problems inherent in air sampling data analysis mean that extreme care must be emphasised in the interpretation of these data. However, even with expert interpretation, it must be recognised that the low-tech high-volume air sampling described in this document is insufficient to detect low but nevertheless hazardous levels of alpha-emitting radionuclides. Reliance on these techniques runs the risk of exposing personnel to unacceptably large radiation doses, potentially up to 200 times greater than would routinely be allowed in the case of an atomic radiation worker.

Haslip, D.S. and Estan, D. 2001. Radiological Air Sampling for the Canadian Forces. DREO TM 2001-033. Defence Research Establishment Ottawa.

Sommaire

Introduction: Le projet 00002199 (détection, identification et dosimétrie de rayonnement) obtiendra sous peut, de l'équipement d'échantillonnage d'air pour les forces canadiennes. Le projet a fait la demande à CRDO d'étudier des méthodologies pour l'échantillonnage d'air et l'analyse de données. Ceci est la rapport final de cette étude, qui a inclus une recherche de la littérature et une série d'essais expérimentales pour démontrer et éprouver les principes qui seront discuté dans ce document.

Résultats: Cet article décrit un protocole pour l'échantillonage d'air et l'analyse de données pour les forces canadiennes. Nous croyons que les forces canadiennes pourraient facilement mettre en application ces méthodes. Nous avons également précisé qu'il existe des imperfections dans cette méthode, particulierement l'effet des produits radioactifs de radon et de thoron empêchant la détection de petites quantités de contaminants suspendus dans l'air. Les méthodes pour éliminer l'effet du radon et du thoron peuvent causer des erreurs car elles sont extrêmement susceptibles aux fluctuations statistiques dans les données. C'est donc l'effet du radon et du thoron qui limite l'efficacité de cette méthode pour la détection de petites quantités de contaminants suspendus dans l'air.

Importance: Les problèmes qui existent dans l'analyse de données d'échantillonage d'air exigent une interpretation méticuleuse de ces données. Cependant, même lorsque l'interprétation est effectuée par un expert, il est important de comprendre que l'échantillonage d'air tel que décrit dans ce document, peut être insuffisant dans la détection de niveaux dangereux d'émetteurs alpha. Dans de telles situations, si trop de confiance est placée sur les techniques décrites dans ce document, il existe un risqué réel d'exposition à de niveaux élevés de rayonnement; même jusqu'à 200 fois plus grandes qu'avec des employées des installations nucléaires.

Haslip, D.S. and Estan, D. 2001. L'échantillonage radiologique d'air pour les forces canadiennes. DREO TM 2001-033 Centre de recherche pour la défense Ottawa.

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1. Introduction

In general, the hazard to personnel from internalised radionuclides is much greater than the hazard due to such sources outside the body. The outer 0.07 mm of the skin is composed of dead cells that are not affected by radiation. This tends to decrease the hazard from beta radiation (especially low-energy radiation) and makes external alpha irradiation harmless. Inside the body, however, no such dead layer exists. For internal exposures, therefore, both alpha and beta radiation present serious hazards.

The three key routes by which radionuclides may be internalised are inhalation, ingestion, and through breaks in the skin. However, whereas external exposure is easily quantified through the personal dosimetry, performing the same function for internal exposures is non-trivial. There are essentially two methods to quantify internal exposures[1]:

1. Surface contamination monitoring - swipes or alpha-beta radiation detectors can be used to detect the presence of radioactive contamination, and therefore the threat of internal exposure. Quantifying exposure from such measurements is very difficult, and requires a number of assumptions about how the radionuclides travel from the surface into the body.

2. Air sampling - this is essentially the only way to determine the concentration of radionuclides in the air, and is thus the only reliable way to estimate inhalation exposures.

Given the danger inherent with internalised radionuclides, it may be surprising that so few air samplers are present in and around establishments holding radioactive materials. The reason for this is that radioactive materials at such facilities are normally well controlled. If one uses only sealed sources, for instance, there is essentially no danger of airborne radioactivity and thus there is no reason to perform air sampling. However, in the military milieu, where uncontrolled sources may be the norm rather than the exception, and where there is the possibility of explosive dispersal, air sampling must be an integral part of the standard procedures.

Although the methods of air sampling are normally very simple, the interpretation of the data can be complex. Indeed, it may be difficult to detect the presence of highly radiotoxic materials in the presence of naturally-occurring airborne radionuclides, such as radon and its daughters. It is the goal of this paper to describe the interpretation of air sampling data, and to delineate how air sampling can be employed to the greatest effect by the Canadian Forces in military operations.

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[1] These methods are often used not to quantify exposures, but rather to provide a warning of contamination, so that exposure can be prevented through the use of appropriate protective measures. However, the task of providing a warning is equivalent to that of straight quantification, because one should first attempt to quantify the exposure before asking if the exposure (which may not have been detectable) is smaller than the warning threshold.

2. Theory of Air Sampling

2.1 Basic Theory

Before delving into the complications inherent in air sampling, it seems prudent to outline the essentials of radiological air sampling in the simplest case. This is done below.

An air sampler passes a volume V of air through a high-efficiency filter (in the case of particulates) or charcoal cartridge (in the case of radio-iodines). The filter or cartridge captures a large fraction F of the radiological substance; one can often assume that this fraction is 1 (refer to the specifications for the sampler filter). Thus, if the airborne radiological concentration (in Bq/m^3, for instance) is C, then the activity collected on the filter will be equal to CVF. This expression assumes that the radioactive substance has a half-life much longer than the sampling time. If it did not, then an appreciable fraction of the activity would decay away during the sample collection.

When a radiation detector counts this activity, the (background-subtracted) count rate R will be equal to the activity multiplied by the detector efficiency D. This detection efficiency (in units such as cps/Bq) takes account of the response of the detector to radiation, the detection geometry, and even any self-absorption of alpha radiation in the filter. It will be isotope-dependent. Thus, the count rate is given by

$$R = CVFD\,.$$

This equation can be rearranged to give the concentration in terms of the other variables:

$$C = \frac{R}{VFD}\,.$$

This concentration can then be used to calculate inhalation doses for personnel without respiratory protection. Given a breathing rate (assume 1.5 m^3/h for moderate exertion) and a stay time in the area, one can calculate the activity that has been inhaled. At this point, one can use tables of "inhalation dose coefficients" [1,2,3] to convert inhaled activity into committed effective dose on an isotope-by-isotope basis. These coefficients vary widely, from tens of picosieverts per Becquerel (for tritium and some iodine isotopes) up to tens of microsieverts per Becquerel (for some neptunium and plutonium isotopes).

2.2 Radon and Thoron

The chief complication in quantifying low-level concentrations of airborne radionuclides is the presence of naturally occurring radionuclides. In air sampling, the problematic isotopes are the daughters of the radon isotopes in the uranium and thorium decay chains.

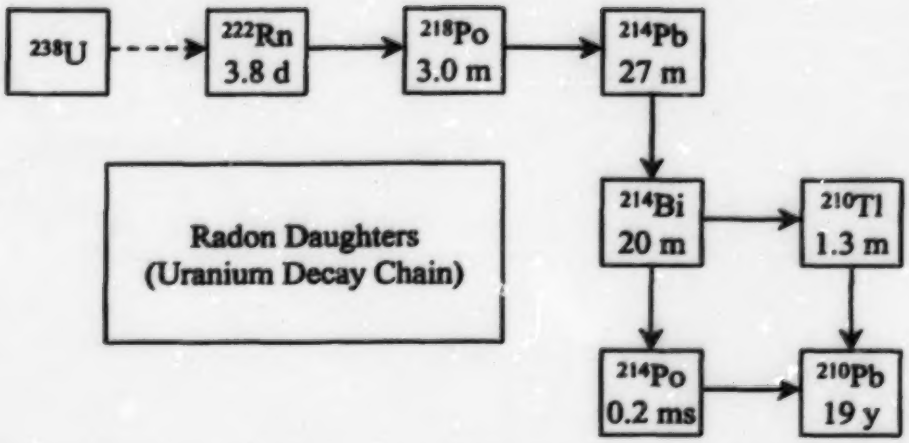

Figure 1. The "radon daughter" part of the uranium decay chain. In this decay table, alpha decays are shown as horizontal arrows, beta decays by vertical ones. Half-lives are shown below the isotope name.

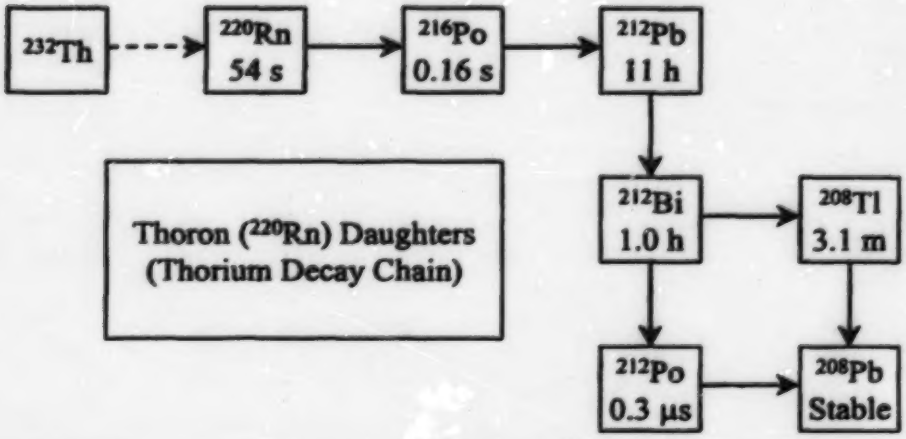

Figure 2. The thoron daughter part of the thorium decay chain. Note the parallels between this chain and the preceding one.

The relevant portions of the ^{238}U and ^{232}Th decay chains are shown in Figure 1 and Figure 2. Both chains pass through an isotope of radon, which is a noble gas. To distinguish between the two isotopes, ^{222}Rn is referred to as "radon", whereas ^{220}Rn (in the thorium chain) is referred to as "thoron".

Because they are gases, radon and thoron are able to escape from whatever solid material they imbue and become part of the surrounding atmosphere (remember that uranium and thorium are present as trace elements in a host of everyday materials, particularly in rocks, soil, and concrete). Averaged world-wide, radon and thoron concentrations outdoors are each around

10 Bq/m^3; indoors, radon concentrations are approximately 40 Bq/m^3, while thoron concentrations are 3 Bq/m^3 [4]. As gases, they will pass through particulate air filters. However their radioactive daughters, formed in the air, attach themselves to dust particles and can thus be trapped in particulate filters.

Since all of the daughters down to ^{210}Pb and ^{208}Pb are relatively short-lived, they should all be present in equilibrium in a given sample of air. The aggregate alpha and beta activity of this host of isotopes can overwhelm the signal from low-level airborne contaminants. The remainder of this section is devoted to exploring methods of eliminating this background from air sampling results. This will help to eliminate false alarms from samples that contain nothing but radon daughters. It will also help to quantify the activity from other contaminants, distinct from that of radon and thoron.

2.3 Radon and Thoron Correction

2.3.1 Theoretical Approach

As we have seen above, the radon and thoron daughters are all relatively short-lived. As a result, the activity on the filter of an air sampler will decrease with time following a sampling. The decay is not exactly exponential, since we are examining a composite of many isotopes with different half-lives, but we can assume for now that the decay is exponential. The composite half-life of radon daughters is found to be about 35 minutes, whereas the thoron daughters' half-life is approximately 10.6 hours [5].

Figure 3 shows a hypothetical measured count rate over 24 hours, assuming equal initial contributions from radon daughters, thoron daughters, and a long-lived unknown isotope. It shows clearly how the total drops quickly at first, reflecting the rapid decay of the radon component, then falls off much more slowly as the longer-lived thoron daughters decay away. The question is how to extract the long-lived component from the total when the individual quantities and ratios are not known.

The generic solution is actually fairly simple, given what is known. The total count rate $C(t)$, as a function of time, is given by the expression

$$C(t) = \mathbf{X}_0 \exp(-\frac{t \ln(2)}{t_{1/2-X}}) + \mathbf{Rn}_0 \exp(-\frac{t \ln(2)}{t_{1/2-Rn}}) + \mathbf{Th}_0 \exp(-\frac{t \ln(2)}{t_{1/2-Th}})$$

where the zero-subscripted quantities are the initial activities of the unknown, radon and thoron, and the denominators in the exponents are the half-lives of the three substances (assume that the half-life of the unknown is very long so the exponential is unity). One could easily perform a fit of this equation to experimental data of the type depicted in Figure 3, and extract the initial activities of all three quantities. In fact, given a sufficiently sophisticated fitting algorithm, one could also fit for the half-lives as a check

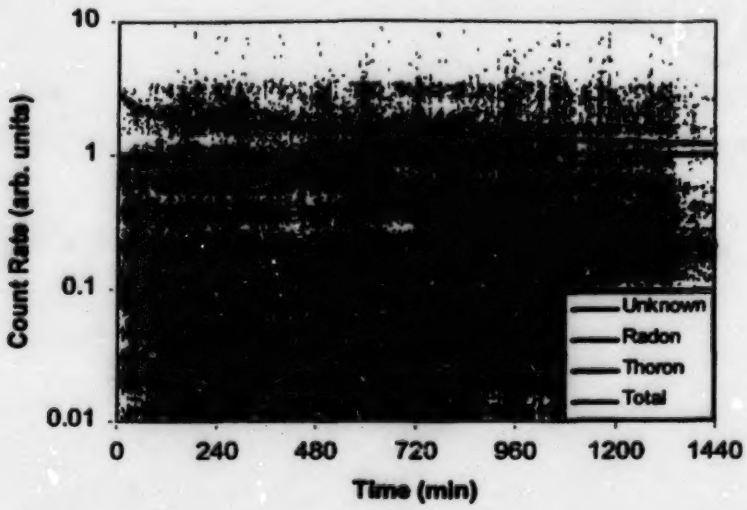

Figure 3. Count rates from radon daughters, thoron daughters, and an unknown long-lived isotope, assuming the contributions from the three are initially equal. The total count rate is also shown.

that the form of the data is as expected. That is, if the fit implied half-lives much different from what is expected, then there is a problem with the data.

2.3.2 Multi-Point Calculations

The above method is correct, but is also ungainly and unreasonable for a field application. Fortunately, a much simpler option is available. In the equation above, there are three unknown quantities (the activities of the three components). Thus, if count rates are measured at three different times following sampling then one can extract the three unknown quantities using the methods of linear algebra. For instance, measurements were taken immediately after sampling, and then 2 and 4 hours later, then the count rates should be

$$C(0) = \mathbf{X}_0 + \mathbf{Rn}_0 + \mathbf{Th}_0$$
$$C(2) = \mathbf{X}_0 + 0.093\mathbf{Rn}_0 + 0.88\mathbf{Th}_0$$
$$C(4) = \mathbf{X}_0 + 0.0036\mathbf{Rn}_0 + 0.77\mathbf{Th}_0$$

where we assume that the unknown's decay is negligible. We can easily reduce this to a system of two equations in two unknowns by subtracting the latter two equations from the first as follows

$$C(0) - C(2) = 0.907\mathbf{Rn}_0 + 0.12\mathbf{Th}_0$$
$$C(0) - C(4) = 0.9914\mathbf{Rn}_0 + 0.23\mathbf{Th}_0$$

Finally, these two equations can be solved in terms of the unknowns, and the solution substituted into one of the previous equations to get the initial activity of X. These expressions are

$$\mathbf{Rn}_0 = 1.23C(0) - 2.64C(2) + 1.40C(4)$$
$$\mathbf{Th}_0 = -0.97C(0) + 11.40C(2) - 10.43C(4)$$
$$\mathbf{X}_0 = 0.74C(0) - 8.76C(2) + 9.02C(4)$$

Thus, as initially stated, all three initial activities are calculated in terms of three measurements. We shall call this kind of calculation a multi-point calculation to remind us that it requires measurements to be made at three distinct times.

More generally, if measurements are made at the conclusion of sampling, and at two later times (call these measurements *C1*, *C2*, *C3*), then the initial activities of the three components are

$$\mathbf{Rn}_0 = RnM1 \times C1 + RnM2 \times C2 + RnM3 \times C3$$
$$\mathbf{Th}_0 = ThM1 \times C1 + ThM2 \times C2 + ThM3 \times C3$$
$$\mathbf{X}_0 = XM1 \times C1 + XM2 \times C2 + XM3 \times C3$$

These coefficients are graphed in Figure 4, Figure 5, and Figure 6 for the cases where the second measurements is taken at 30 minutes, 1 hour, and 2 hours after sampling. These values are also tabulated in Appendix A. The important point to note is that when the coefficients are large, then small uncertainties in measurements can translate into large uncertainties in radiological concentrations. The figures show that the best way to keep the coefficients small is to make the three measurements at times that are "significantly different" in terms of the radioactive decay. For instance, making three measurements in rapid succession following sampling is useless, since it provides very little information about the thoron concentration as opposed to the long-lived contaminant. Likewise, making all measurements at days after the sampling says nothing about the radon concentration. The optimal situation is to make one measurement when the sample is completed, a second at least 30 minutes later (when an appreciable quantity of radon has decayed away), and the last after 10 hours (when an appreciable quantity of thoron has decayed away). Since this timeline probably conflicts with operational tempo, the last time can be decreased somewhat, with concomitant increases in uncertainty.

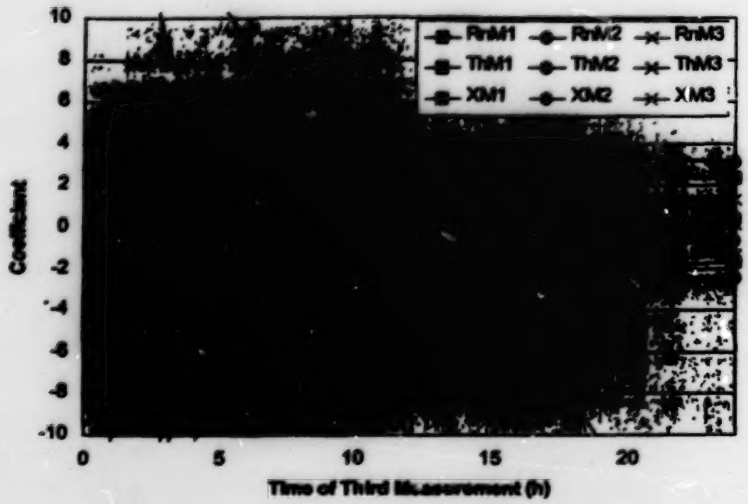

Figure 4. Multi-point calculation coefficients for the case when the first measurement is taken at the conclusion of the sample and the second is taken after 30 minutes.

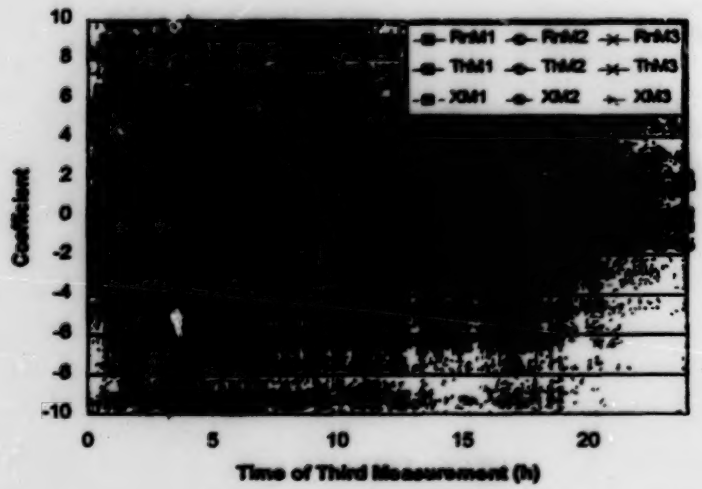

Figure 5. Multi-point calculation coefficients for the case when the first measurement is taken at the conclusion of the sample and the second is taken ater 1 hour.

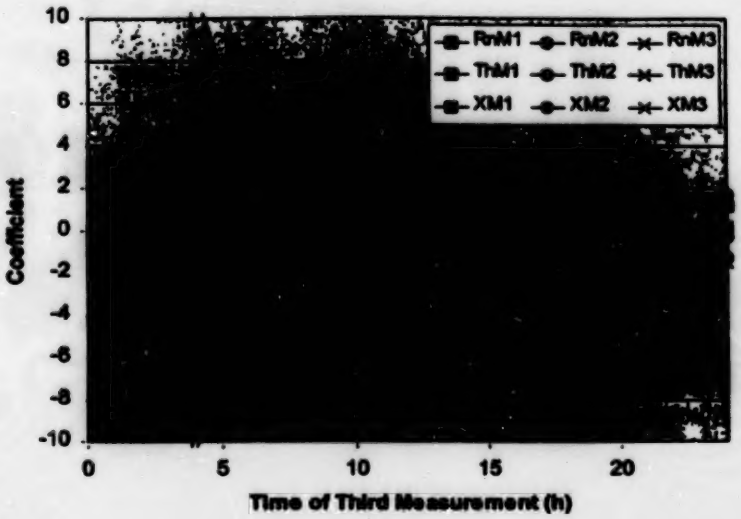

Figure 6. Multi-point calculation coefficients for the case when the first measurement is taken at the conclusion of the sample and the second is taken after 2 hours.

2.3.3 Shortcuts

In the field, it may not always be practical to wait four hours, or even thirty minutes, for the results of an air sample. The following techniques may be used to get approximate results or quick indications in an expedient fashion.

2.3.3.1 *Neglected or Partial Background Subtraction*

The simplest technique to eliminate the complications of radon-thoron subtraction is not to do it. That is, assume that there are no radon or thoron daughters on the filter, and assume that all counts are due to a radiological contaminant. Note that this should only be used if one is sure that there is a contaminant, and not if the air sampling is being used as a warning tool. Confirmation of the hazard must come from another method.

If one can afford to wait for 35 minutes, a good alternative is to assume that all of the background counts come from radon, as opposed to thoron. After 35 minutes, half of the thoron daughters should have decayed away. Thus, taking the original count rate and subtracting twice the difference between the original count rate and the count rate after 35 minutes yields the radon-corrected count rate. For instance, if the initial count rate was 20 cps, and the count rate at 35 minutes was 16 cps, then the radon-corrected

count rate is 12 cps. As in the previous case, this method cannot be used as a first alert, since the result will be non-zero in the presence of thoron daughters.

A third option in this vein is to subtract background count rates measured at other locales or at this locale at earlier dates. This method, too, can be very problematic, since radon and thoron concentrations can be extremely sensitive to location, and even time of day (radon and thoron concentrations are normally highest in the morning and fall throughout the day).

2.3.3.2 *First-Count Factor*

This is a way of quickly determining the presence, if not the quantity, of a non-radon, non-thoron contaminant in the air sample. The premise is simple. Radon daughters emit both alphas and betas, so alpha detectors and beta detectors will both measure radiation on sampler filters. Regardless of the quantity of radon daughters in the atmosphere, the ratio of count rates measured by the two detectors should be a constant [5]. Moreover, since the decay scheme of thoron is almost identical to that of radon, the alpha-beta detection ratio for thoron daughters should be the same as that for radon daughters. However, any other contaminant will have a different proportion of alphas and betas (and probably all of one and none of the other). Thus, one can acquire an air sample, measure the alpha-beta ratio, and compare to that on an earlier, contaminant-free sample. A difference signals the presence of a contaminant.

2.3.3.3 *Gamma Spectroscopy*

Gamma spectroscopy is an effective method for unambiguously identifying a wide variety of radioactive isotopes. It can and should be used on contaminated air sample filters to identify what the contaminant is, which is important in choosing the inhalation dose coefficient. However, it could also be used on filters whose status was unknown, in order to determine if a contaminant is present. This could be very sensitive to low-level contaminants, but will not help with isotopes that emit only alpha, beta, or low-energy gamma rays.

2.3.4 Short-Lived Isotopes

Things become considerably more complicated when the contaminating isotope is short-lived, rather than long-lived, as we have assumed up to now. First, as described earlier, the activity on the filter after a sample is not *CVF*, but rather [5]

$$\frac{CVFt_{1/2}}{t\ln(2)}\left(1-\exp\left(\frac{-t\ln(2)}{t_{1/2}}\right)\right).$$

Incidentally, this equation means that the quantity of radionuclide that can be captured has a maximum (it does not grow without bound) and as a result, the amount that is captured is smaller than expected by an amount that increases with time. Practically speaking, it doesn't make sense to sample for longer than 2 or 3 half-lives [5].

Next, all of the multi-point calculations derived earlier are invalid and must be re-derived. Further, when the half-life of the contaminant approaches that of radon or thoron, it becomes impossible to subtract the background without resorting to a more advanced technique, such as gamma spectroscopy or a more sophisticated application of the first-count principle. Fortunately, isotopes with short half-lives do not have large inhalation dose coefficients (if the half-life is less than a day, then the inhalation dose coefficient will be smaller than about 1 nSv/Bq). Thus, if one is forced not to subtract background, the uncertainty introduced is not as large.

3. Air Sampling in Practice

Air samples were taken and measured at DREO in a number of different locations and with a number of different sampling parameters. This section of the document highlights the outcomes of this work with the goal of demonstrating some of the principles outlined in the previous section. Important to note are the difficulties inherent in obtaining a reliable measurement of airborne radionuclides.

3.1 An Example Background

Figure 7 shows the alpha and beta count rates measured from an air filter following a one-hour sample at a flow rate of 100 litres/minute in room 29 at DREO. It shows that both rates fall off in approximately the same way. The lines are fits of exponential functions to these data sets, and allow one to measure the half-life of the decay. These half-lives are 43 minutes in the case of the alphas and 31 minutes in the case of the betas. The average is 37 minutes, very close to the 35-minute half-life of radon given in the preceding section. The important point to note is that there is no evidence for long-lived thoron daughters in these data, or in any of the measurements taken at DREO. This is somewhat unfortunate, but points out the variability in the relative quantities of the two gases (since we expect to see equal quantities of the two in at least some of the samples taken in this work).

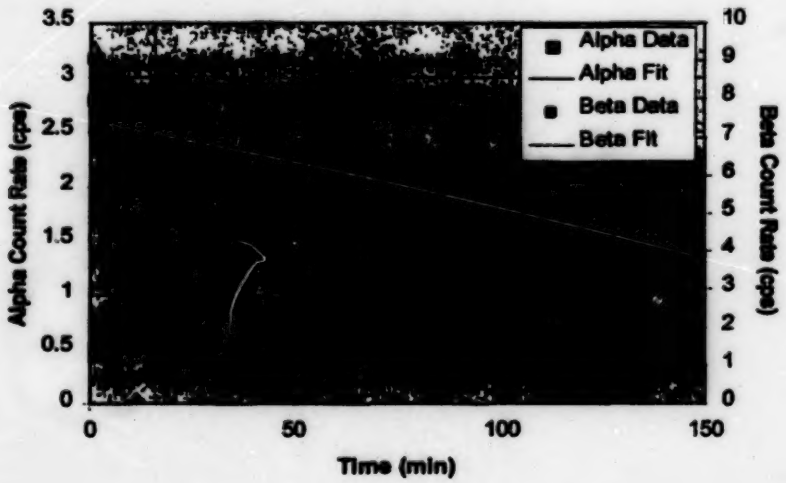

Figure 7. Count rates on an air filter following a one-hour sample at DREO. Squares show the background-subtracted count rates measured by an ABP-100 alpha-beta probe; lines show regression fits to these data.

3.2 Temporal Variation

It was earlier noted that radon and thoron concentrations can vary significantly with time of day and with location. This section looks at the first of these factors. Figure 8 shows the count rate on air sampler filters following five one-hour samples at five different times during the day, all in the same room in the basement of Building 5B at DREO. As expected, the radon concentration is greatest in the morning, falling steadily as the day progresses. The figure shows that the morning:afternoon ratio is approximately 2:1. In fact, it can be much larger. A measurement taken in this room the next morning yielded a count rate of 35 cps, making the radon concentration 7 times larger than it had been only 17 hours earlier. This highlights the problem with subtracting previously measured background radon concentrations, as discussed in Section 2.3.3.1.

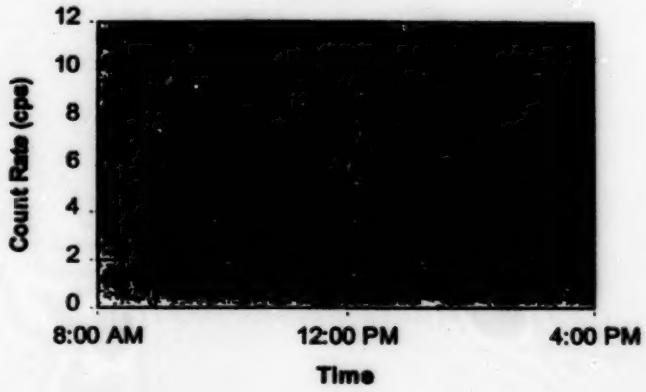

Figure 8. Count rates following one-hour air samples at various times throughout the data at a given location at DREO. The count rates are summed alpha-beta rates measured with a Liquid Scintillation Counter, and are averaged over the first 20 minutes following the sample.

3.3 Spatial Variation

Figure 9 shows the corresponding spatial variation in radon concentrations. The figure shows the alpha count rates, as a function of time, on air sampler filters following air sampling at four different locations at DREO (all measurements were made in the morning). The rates differ by as much as a factor of 15 between different locales. It is worth noting that once again we have performed exponential fits on these data to extract half-lives. The half-lives for the Building 29, Building 24, Building 5B, and Outside data are 43 minutes, 67 minutes, 42 minutes, and 32 minutes, respectively. Based on the variation we saw earlier in this kind of calculation, all but the second measurement are easily consistent with the expected value of 35 minutes. In the last case, it is tempting to believe that the long half-life is indicative of the presence of thoron daughters. However, this does not stand up under scrutiny. If one restricts the fit to the first 30 minutes of data, when the radon signal should be most dominant, the fit indicates a half-life of 71 minutes. That this doesn't decrease when one restricts the fit

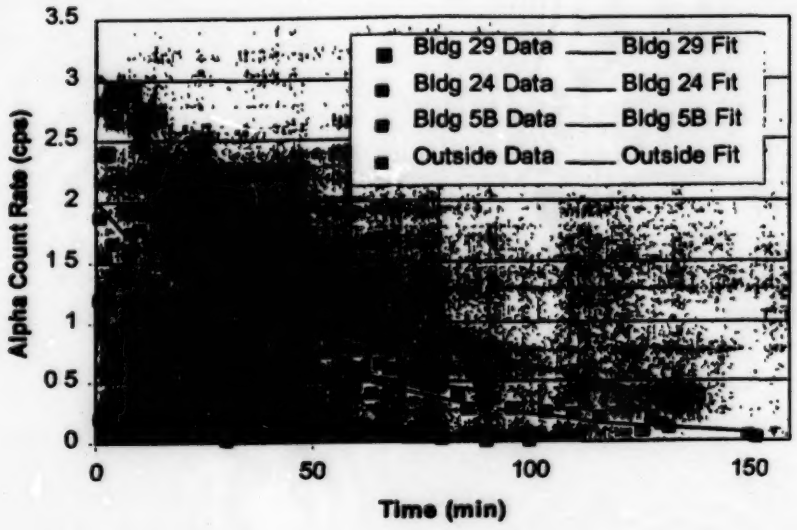

Figure 9. Alpha count rates on air sample filters following one-hour 60 lpm air samples at four locations around DREO.

indicates that the erroneous value derives from problems in the data (inadequate background subtraction, large fluctuations) rather than the presence of thoron.

3.4 Applying Radon Correction Methodologies

In the process of performing this work, 13 air samples were taken and the filters counted. This section uses the radon correction methodologies of Section 2.3 on these data to see how successful they are on a blank (background) sample. Table 1 summarises these results for the 13 runs. It shows the first-count factor, and the results of the quick method (based on measurements made immediately after sampling and then at 35 minutes) and a multi-point calculation, using both alpha and beta count rates. The last column is a "Comments" field that summarises the results of the quick and multi-point methods. These are colour-coded green, yellow, and red according to whether the results were good, mixed, or poor, respectively.

The first-count factor averages 0.42±0.17; more practically, it should be characterised as 0.4 within a factor of two. This is not a very sensitive measure for the presence of contamination, but it may prove more useful than some of the other methods, especially if the contaminant exclusively emits only one form of radiation. For instance, uranium may not be detectable with this method since uranium and its daughters emit a combination of alphas and betas.

With the prediction that non-radon contaminants make up more than 25% of the original reading in 11 out of 24 (46%) cases (when this is never the case), the partial subtraction method is successful only about half of the time. As discussed earlier, this method is very

Table 1. Results of using some of the various background-subtraction methodologies of Section 2.3. "Partial Subtraction" indicates the quick method where the measurements are made immediately after sampling and again at 35 minutes. Both Partial Subtraction and Multi-point results are given in terms of a percentage of the initial count rate. The comments field summarises the results and is colour coded according to whether the result is a success (green), marginal (yellow), or a failure (red).

Run	First-Count Factor	Partial Subtraction	Multi-point Calculation	Comments
1	---	---	---	Measurements negative
		96%	-5751%	MPC right, PS wrong
2	0.32	-65%	-2148%	Both predictions negative
		28%	1878%	MPC blows up
3	0.21	44%	-2325%	MPC right, PS wrong
		-12%	1825%	MPC blows up
4	---	---	---	Measurements negative
		-55%	2223%	MPC blows up
5	0.59	-96%	468%	MPC blows up
		9%	-50%	Predictions low to negative
6	0.74	103%	-5294%	MPC right, PS wrong
		52%	-2741%	MPC right, PS wrong
7	0.40	-92%	5773%	MPC blows up
		51%	-610%	MPC right, PS wrong
8	0.64	-27%	3460%	MPC blows up
		15%	71%	MPC indicates non-zero value
9	0.27	32%	162%	Both indicate non-zero value
		61%	-516%	MPC right, PS wrong
10	0.40	36%	-1858%	MPC right, PS wrong
		18%	1190%	MPC blows up
11	0.41	25%	-80%	MPC right, PS low
		51%	-926%	MPC right, PS wrong
12	0.33	22%	-126%	MPC right, PS low
		23%	-204%	MPC right, PS low
13	0.31	17%	-136%	MPC right, PS low
		42%	-423%	MPC right, PS wrong

primitive, and can be lead astray by statistical fluctuations in the measurements, as demonstrated here.

The multi-point calculations fared little better, predicting positive (and significant) levels of non-radon contamination in 9 out of 24 (38%) cases. All of these cases used a second measurement at 30 minutes following sampling. In addition, most used a third measurement at 1 hour after sampling. Cases 5, 9, 12, and 13 used third measurements at longer times (5 hours in the first case, 2 hours in the rest). This shows in the better success ratios (25% versus 44% for the 1-hour results) and in the less outlandish predictions (none in the thousand-percent range). In fact, one can see that the multi-point calculation with the third measurement at 1 hour post-sample is less effective than the primitive method reviewed above, having the same false positive rate, and producing a slew of unbelievable results. Even

the two-hour multi-point calculations are uncertain and unreliable, but not to as great an extent.

One of the problems with these data is clearly the amount of statistical fluctuation that is present. This is characteristic of the ABP-100 probe when it is operated in survey mode. One way around this is to measure the count rates over longer times (say, two minutes); this is accomplished with the ABP-100 in Scaler mode. Reduction in fluctuations will help to eliminate some of the more outlandish results seen above. However, it does not negate the inherent sensitivity of the multi-point calculations to non-ideal data. The importance of having the third data point at least two hours post-sampling, and of using good judgement in assessing the results of such calculations cannot be overstated.

4. Recommendations and Conclusions

4.1 Sampling Method

One of the goals of this Engineering Study was to provide guidance to DND on how to use air sampling in the field. The following sampling methodology is based on the research undertaken as a part of this study, as well as the experience gained from the experimental work undertaken in this study.

High-volume air samplers are typically capable of sampling air at flow rates exceeding 100 litres per minute. For the applications of interest here, there is no reason to take samples at low flow rates. Thus, the sampler should be operated at a high flow rate; 100 litres per minute is a good rate from the standpoint of computational complexity.

One hour is a reasonable sampling time, and will be used in the sensitivity calculations that follow. Ultimately, shorter sampling times are acceptable if you can work with the reduced sensitivity. On the other hand, if one wants higher sensitivity, one can increase the sampling time. It should be noted that one cannot get arbitrarily large sensitivity simply through increasing the sampling time. Finally, if the isotope of interest has a short half-life, then one shouldn't sample for longer than 2 or 3 half-lives. Longer sampling times will bring very little gain, and may even bring very large backgrounds.

Once the sample is taken, an alpha-beta probe (such as the NRC ABP-100) should be used to measure the activity on the filter. Although this is probably not as sensitive as is desirable, it is the best field-expedient method. It also has the advantage that it is non-destructive, meaning that the filter can be retained for further analysis, or as evidence if required. The measurement should be made with a minimum distance between the probe and filter, in a reproducible geometry. For the counting, average count rates over two minutes should be used, in order to reduce fluctuations (with the ABP-100, this can be done with the Scaler mode). Both alpha and beta count rates must be measured separately. Measurements should be taken (a) immediately after sampling, (b) thirty minutes later, and (c) at least 2 hours after the original measurement. It would be best to have a series of measurements at half-hour intervals (0 minutes, 0.5 hours, 1 hour, 1.5 hours, 2 hours, ...).

Both the partial subtraction and multi-point calculations should be used as in the text to estimate the non-radon activities. As this work has shown, neither of these methods is particularly reliable, and so no result should be viewed as unassailable. Clearly, any very large numbers emerging from the multi-point calculations should be viewed as suspect.

Gamma-ray spectroscopy should be used to analyse all filters, particularly in view of the inaccuracies of the other analysis methods. Gamma spectroscopy can be useful in detecting the presence of low-level contaminants, and to identify these substances. However, spectroscopy is not capable of detecting all possible hazards, and a negative result from a gamma spectroscopy system is not the final word. All of the methods discussed in this section must be used in concert, and expert guidance will probably be necessary.

In general, it is difficult to improve one's data through the use of previous background measurements. Spatial and temporal variations in radon and thoron concentrations will likely mitigate the effectiveness of such measurements. The only way in which they may be useful is as an indicator of whether long-lived thoron is present, or whether the background is entirely radon (as observed in this work). The presence of thoron will undoubtedly pose difficulties for background subtraction, since its slow decay will be difficult to differentiate from the constant activity of a long-lived contaminant.

It would be useful to validate this sampling methodology with actual radioactive contamination. The possibility for such trials should be investigated once 00002199 has decided on an air sampler.

4.2 Sensitivity

Based on the knowledge acquired during this study, one can estimate the sensitivity of this methodology to airborne contaminants. Calculations of this sensitivity are summarised in Table 2, and will be explained in this section.

This work has demonstrated the difficulty of subtracting the radon backgrounds from air sampling data. Thus, we have conservatively estimated that a radioactive contaminant would have to produce 3 cps of alpha activity or 10 cps of beta activity to be detectable. These values are taken from Figure 7, which is the data set in which the highest levels of radon activity were seen. It is certainly possible that with careful work, levels lower than this could be seen. It is also true that this is a worst case in terms of radon levels seen in this study, and that as the background drops, so too does the detectable amount of contaminant. However, it should also be noted that the CF may need to perform sampling in areas where the background is much higher. In addition, the air samples collected in this study were devoid of thoron; if thoron was present, its longer (10 h) half-life could pose serious difficulties for

Table 2. Sensitivity of the air sampling method to various radioisotopes. The first four isotopes are alpha emitters (although there may be beta emitters in the decay chain), whereas the second group of four are all beta emitters. The annual dose in the rightmost column assumes 24 hour a day exposure.

Isotope	Sensitivity (cps/(Bq/cm^2))	Areal Activity (Bq/cm^2)	Activity (Bq)	Volumetric Activity (Bq/m^3)	Inhalation Dose Rate (mSv/h)	Annual Inhalation Dose (mSv)
^{234}U	0.26	11.5	236	39.3	0.206	1810
^{241}Am	0.57	5.26	108	17.9	1.13	9890
^{238}U	3.5	0.86	17.5	2.92	0.0127	111
^{232}Th	8.3	0.36	7.38	1.23	0.818	7160
^{14}C	0.025	400	8170	1360	0.00409	35.8
^{99}Tc	0.44	22.7	464	77.4	2.61E-04	2.3
^{36}Cl	2.2	4.55	92.9	15.5	1.38E-04	1.2
^{90}Sr	4.4	2.27	46.4	7.74	4.18E-04	3.7

contaminant measurement. Thus the 3 cps and 10 cps values are taken as conservative but not overly pessimistic.

In a previous work [6], the sensitivity of the ABP-100 probe to various alpha- and beta-emitting nuclei was quantified. These values are given in the table, corrected to account for the size of the air sample filter (approximately 5 cm in diameter). These values, combined with the 3 cps/10 cps values discussed above, give the areal activities in the table. These are the minimum levels of contaminant that will be detectable. These range over several orders of magnitude, depending on the characteristics of the radiation in each case. When multiplied by the area of the filter, these give the total activities in column 4. Assuming that these data were taken in a one-hour, 6000-litre sample, we can derive the volumetric activity in the next column. This is the minimum activity concentration that can be detected with this air sampler. As noted above, higher volume air samples will lower this sensitivity somewhat (since the background will likely be better characterised, and in the case of radon will not increase with longer times), whereas lower volume samples will increase this minimum detectable level.

As described earlier in this report, there exist tables of so-called "inhalation dose coefficient", numbers that convert inhaled activities (in Becquerels) into committed effective doses (in Sieverts). We use the volumetric activities in the table and an assumed breathing rate of 1.5 m^3/h to derive the inhalation dose rate in the second-to-last column. This is not an acute dose, but rather is received over an extended period. Nevertheless, it is a radiation dose that carries with it all of the consequences of long-term exposures. It should be noted that the NATO guidance on dose rate (which does not differentiate between external and internal committed exposures) uses 0.002 mSv/h as the hazard perimeter dose rate and 0.003 mSv/h as the turn-back dose rate [7]. One of the beta emitters and all of the alpha emitters exceed these values; in some cases, the value is exceeded by 300-400 times. To state this another way, the air sampling methodology does not allow the CF to detect airborne radioactivity concentrations to a level such that NATO guidance can be obeyed. In some cases, the guidance can be exceeded by enormous factors.

To put this in a different context, these dose rates have been converted to annual doses, assuming 24-hour-a-day exposures. These can be compared to the annual civilian limit of 1 mSv, and the annual average limit for radiation workers of 20 mSv. All of the cases described here exceed the 1 mSv limit, and five of them exceed the 20 mSv level. In the case of ^{14}C, the level is not too much larger, and does not exceed the one-year allowable level of 50 mSv for radiation workers (the average over 5 years must be less than 20 mSv). However, all of the alpha emitter doses exceed this level, and three of them do so by great amounts (factors of 90-500). DND personnel could be in highly dangerous environments and yet not be able to detect, let alone quantify, the hazard. This implies that DND should consider other methods of detecting airborne contaminants in high-risk areas.

Automated alpha-beta air monitors are one such method of reducing this risk. These devices contain internal silicon detectors that are ultra-sensitive and capable of subtracting radon activity through alpha spectrometry. This gives them sensitivities in the hundredths of Bq/m^3, bringing the inhalation doses down to more acceptable levels (the exact level depends on the device and the isotope). Of course, these high-technology items are less rugged and require more training to use, but would be well worth having under certain circumstances.

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Appendix A: Multi-Point Calculation Coefficients

Table 3. Multipoint calculation coefficients for the case when the first measurement is taken at the conclusion of the sampling, and the second is taken 30 minutes later. T_3 is the time of the third measurement.

T_3 (h)	RnM1	RnM2	RnM3	ThM1	ThM2	ThM3	XM1	XM2	XM3
1	5.195	-10.563	5.367	-41.417	116.441	-75.025	37.221	-105.879	69.657
2	3.513	-4.762	1.249	-17.901	35.365	-17.464	15.388	-30.602	16.214
3	3.007	-3.669	0.662	-10.825	20.084	-9.259	8 818	-16 415	8.596
4	2.780	-3.231	0.451	-7.655	13.965	-6.310	5.875	-10.734	5.859
5	2.657	-3 003	0.346	-5.935	10.775	-4.839	4 278	-7 771	4.493
6	2.582	-2 866	0.284	-4.882	8 850	-3.967	3 301	-5 984	3.684
7	2.531	-2.774	0.243	-4.180	7.574	-3.394	2.649	-4 800	3.151
8	2.496	-2.710	0 214	-3.682	6.671	-2.988	2.186	-3.961	2.775
9	2.469	-2 662	0 192	-3.312	6.000	-2.688	1.843	-3.338	2.495
10	2.449	-2 625	0.176	-3.027	5.483	-2.456	1.578	-2.858	2.280
12	2.420	-2 572	0 152	-2.617	4 741	-2.124	1.198	-2 170	1.972
14	2.400	-2 536	0.136	-2.340	4.238	-1.898	0.940	-1 702	1.763
16	2.385	-2.510	0 124	-2.140	3.877	-1.737	0.755	-1 367	1.612
18	2.375	-2.490	0.116	-1.991	3.607	-1.616	0.616	-1.117	1 500
20	2.367	-2.476	0.109	-1.877	3.399	-1.523	0.510	-0.924	1.414
22	2.360	-2 464	0.104	-1.786	3.236	-1.449	0.426	-0.772	1.346
24	2 355	-2 454	0.099	-1.714	3.105	-1 391	0.359	-0 650	1.291

Table 4. Multipoint calculation coefficients for the case when the first measurement is taken at the conclusion of the sampling, and the second is taken 1 hour later.

T_3 (h)	RnM1	RnM2	RnM3	ThM1	ThM2	ThM3	XM1	XM2	XM3
2	2 132	-4.407	2.275	-7.644	32.725	-25.081	6.512	-28.318	22.806
3	1.842	-2.857	1.015	-4.449	15.637	-11.188	3.607	-12.780	10.174
4	1.715	-2.366	0.650	-3.054	10.224	-7.170	2.339	-7.858	6.520
5	1.649	-2.132	0.484	-2.317	7.650	-5.333	1.669	-5.518	4.849
6	1.609	-1 998	0.390	-1.877	6.171	-4.294	1.269	-4.173	3.904
7	1.583	-1.912	0.329	-1.590	5.219	-3.630	1.007	-3.308	3.300
8	1.564	-1 852	0.288	-1.389	4.559	-3.170	0.825	-2.707	2.882
9	1.551	-1.808	0 257	-1.242	4.076	-2.834	0.691	-2.268	2.577
10	1.541	-1 775	0 234	-1.130	3.707	-2.577	0.589	-1.932	2.344
12	1.526	-1.727	0.201	-0.970	3 184	-2.214	0.444	-1.457	2.013
14	1.517	-1 695	0.179	-0.864	2 834	-1.970	0 347	-1.138	1.791
16	1 510	-1.673	0.163	-0.787	2.584	-1.796	0.278	-0.911	1.633
18	1.505	-1.656	0 151	-0.731	2.398	-1.667	0.226	-0.742	1.516
20	1.501	-1.643	0.142	-0.688	2.256	-1.569	0.187	-0.613	1.426
22	1 498	-1.633	0.135	-0.654	2 144	-1.491	0.156	-0.512	1.356
24	1.495	-1.625	0.130	-0 626	2.055	-1 429	0 131	-0.430	1.299

Table 5. Multipoint calculation coefficients for the case when the first measurement is taken at the conclusion of the sampling, and the second is taken 2 hours later

T_3 (h)	RnM1	RnM2	RnM3	ThM1	ThM2	ThM3	XM1	XM2	XM3
3	1.308	-4.193	2.886	-1.525	22.953	-21.428	1.217	-18.759	18.542
4	1 233	-2 637	1.404	-0 969	11 397	-10 428	0.736	-8.760	9.024
5	1 196	-2.133	0.937	-0 692	7 652	-6 960	0.497	-5 519	6.022
6	1 175	-1.887	0.713	-0.537	5 829	-5 292	0 362	-3.941	4.579
7	1.162	-1.743	0.582	-0 441	4 759	-4 318	0.279	-3 016	3.737
8	1 153	-1.649	0.496	-0 377	4.060	-3 683	0.224	-2 411	3.187
9	1.147	-1.583	0 436	-0.331	3.568	-3 237	0.184	-1 985	2.801
10	1.142	-1.534	0.391	-0 298	3.204	-2 907	0 155	-1 670	2.515
12	1.136	-! 466	0 330	-0.251	2.704	-2.453	0.115	-1.237	2 122
14	1.132	-1 423	0 290	-0.221	2.378	-2.157	0 089	-0.955	1.866
16	1.129	-1 392	0.263	-0.200	2.150	-1.950	0 070	-0.758	1.688
18	1 127	-1.369	0.242	-0.184	1.983	-1.799	0.057	-0.614	1.557
20	1 126	-1.352	0.227	-0 172	1 857	-1.685	0.047	-0.505	1 458
22	1 124	-1 339	0.215	-0 163	1 759	-1.595	0.039	-0.420	1.381
24	1 123	-1.329	0.205	-0.156	1 681	-1 525	0.033	-0 352	1.319

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DREO has performed a study of air sampling methodologies and methods of data analysis following air sampling. This has been done in support of Project 00002199 (Radiation Detection, Identification, and Dosimetry) and is focussed on what methods would be best suited to the Canadian Forces. As part of this work, DREO performed a number of experimental trials to demonstrate and test the principles and methods of air sampling. A suggested method is presented for how the CF should perform air sampling. It uses high-volume air samplers, alpha-beta probes for detection of contaminants on the filter, and gamma-ray spectroscopy for additional sensitivity in some cases. Data analysis approaches are suggested, with appropriate cautions on their use. One of the major findings of this study is that radon and thoron backgrounds present a serious impediment to the detection of low levels of airborne contaminants. In fact, the equipment discussed in this work is insufficient for the detection of alpha-emitting contaminants, even when the field-optimised methods of this study are employed. Reliance on these techniques in such scenarios risks high-level radiation exposures.

14 KEYWORDS, DESCRIPTORS or IDENTIFIERS (technically meaningful terms or short phrases that characterize a document and could be helpful in cataloguing the document. They should be selected so that no security classification is required Identifiers such as equipment model designation, trade name, military project code name, geographic location may also be included If possible keywords should be selected from a published thesaurus e g Thesaurus of Engineering and Scientific Terms (TEST) and that thesaurus-identified If it is not possible to select indexing terms which are Unclassified, the classification of each should be indicated as with the title.)

radiation
air sampling
radon
thoron
radioactive background

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